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<p>(21) International Application Number: PCT/US90/06575</p> <p>(22) International Filing Date: 9 November 1990 (09.11.90)</p> <p>(30) Priority data: 438,927 17 November 1989 (17.11.89) US</p> <p>(71) Applicant: W.L. GORE & ASSOCIATES, INC. [US/US]; 551 Paper Mill Road, P.O. Box 9206, Newark, DE 19714 (US).</p> <p>(72) Inventor: SAKHPARA, Dilip, J. ; 159 Boyer Drive, Newark, DE 10702 (US).</p> <p>(74) Agents: SAMUELS, Gary, A. et al.; W.L. Gore & Associates, Inc., 551 Paper Mill Road, P.O. Box 9206, Newark, DE 19714 (US).</p>		<p>(81) Designated States: AT (European patent), BE (European patent), CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (European patent), IT (European patent), JP, LU (European patent), NL (European patent), SE (European patent).</p> <p>Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>
<p>(54) Title: THIN SEAM SEALING TAPE</p> <div style="text-align: center; margin-top: 100px;"> </div>		
<p>(57) Abstract</p> <p>Thin layered tapes for sealing textile seams against water entry wherein the first layer is a thermoplastic hot melt adhesive resin, the second layer consists of a microporous expanded polytetrafluoroethylene membrane whose pores are at least partly filled with a substantially transparent and colorless high melting or non-melting polyurethane adhesive. This adhesive binds the thermoplastic layer to the membrane, and optionally a third layer of very thin material to the second face of the membrane when the pores are essentially filled with adhesive. The thinness of the structures provides sealed seams with less bulk, good hand and drape and easier application. The compression and heating of the tape during application further reduces the reflectivity and increases the transparency of the tape on the sewn seam.</p>		

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THIN SEAM SEALING TAPE

FIELD OF THE INVENTION

This invention relates to a layered tape for sealing sewn seams of waterproof fabric to prevent leakage of water or other liquids through the sewn joint and provide sealed seams which have low reflectance and are essentially transparent in the final garment.

BACKGROUND OF THE INVENTION

The need to seal sewn seams to provide waterproofness has been known for many years. Early approaches to solve this problem involved the application of rubber solutions in volatile solvents, or adhesively bound strips of rubber to the seams. Later, commercially available synthetic latex dispersions were used for the same purpose with rubberized cloth. Such methods are still in use.

With the advent of breathable waterproof fabrics, that is, fabrics which repel liquid water but allow the passage of water vapor, and their use in outer garments, the same problem of seam sealing arose again. Initially, welding the outer fabric was tried instead of sewing. Welding was slow and the seams were generally not as strong as sewn seams and it was found the welded seams were not necessarily waterproof. Later, various pastes in volatile solvents were painted on the seams, but this was messy and the rate of failure at the seams was far too high. Seam sealing tapes were adopted to solve the leakage problem.

To provide a durable waterproof seam it is necessary to enclose and seal the sewing thread and the ends of the fabric at the seam with a waterproof material. Sealing is accomplished using a thermoplastic hot melt adhesive. It is necessary to ensure that the hot melt adhesive has penetrated the fabric on both sides of the seam all of the way through the fabric to the material which supplies waterproofness and breathability to the garments. Seam tape is optionally used on either the lining fabric or shell fabric or directly on the material providing waterproofness.

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On the surface of the garment where two seams intersect, it is necessary to have the sealing tape on one seam cross over the sealing tape on the second seam.

The structure of sealing tapes, especially those for use with breathable fabric laminates utilizing expanded porous polytetrafluoroethylene membrane as a breathable liquid barrier, contain at least two layers; the first layer is most commonly a hot melt adhesive and the second layer is a waterproof layer which can be expanded porous polytetrafluoroethylene or a high melting or non-melting film layer. The film may be any of a number of plastic materials.

The choice of materials for the thermoplastic hot melt adhesive is fairly wide. Thermoplastic films of polyester, polyamides, fluoropolymers and polyurethanes are all used commercially, as hot melt adhesives.

Sometimes a third fabric layer is used. The fabric is usually the same fabric both in structure and color as used in the garment lining. This third layer is intended for both aesthetic purposes and to provide abrasion resistance.

Tapes containing an expanded porous polytetrafluoroethylene layer and/or with a third top fabric layer have an advantage of being more dimensionally stable during application than those without the third layer. Application of the tape over a seam involves heating the thermoplastic hot melt adhesive, usually in a directed blast of very hot air, up to 800°C, although the tape does not achieve anything like that temperature. High air temperatures are used to permit the speed of the seam sealing operation to be accelerated and residence time of the thermoplastic exposed at such temperatures is very short. Immediately after being exposed to the hot air, the fabric with the tape over the seam is fed into a compression zone to press the softened thermoplastic adhesive layer into the fabric. The very nature of the operation induces intermittent motion which, if the tape is not stabilized by expanded microporous polytetrafluoroethylene and/or a layer of fabric, tend to stretch and narrow the hot tape.

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Heavier, thicker tapes have several notable problems induced by their bulk and stiffness. Heavier or thicker tapes have a larger bending radius than thinner or lighter tapes, making it difficult in getting a good seal at crossover seam points due to the inability of the thicker tapes to conform to the contours of the edges of the thick tape it is crossing. Heavier and thicker tapes also make bulkier, stiffer seams which detract from the drape and handle of the garment and, even if the tape is applied to the inside of the garment. The bulkier stiffer seams on the inside of the garment result in the seams being high spots when they are put under tension. These high spots are commonly areas of failure if located on portions of a garment that experience wear.

SUMMARY OF THE INVENTION

Thin layered tapes for sealing sewn textile seams to liquid entry are made by adhering an essentially colorless, transparent elastomeric thermoplastic hot melt adhesive layer to a liquid water repelling, water vapor transmitting expanded microporous polytetrafluoroethylene membrane whose pores have been at least partially filled with an essentially colorless, transparent liquid breathable polyurethane prepolymer that is cured to a form a high melting flexible transparent elastomer with good adhesive properties.

Optionally, if the expanded microporous polytetrafluoroethylene membrane is substantially filled with the liquid polyurethane prepolymer a third layer of non-woven, very sheer fabric may be adhered to the second face of said membrane.

The layered tapes of this invention provide sealed seams that are substantially transparent and low in reflectance when heated and put under pressure during application making them almost invisible on a fabric seam. Being very thin, the tapes provide less bulk in the seam, good hand and drape and are easier to apply to the seam than thicker tapes.

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BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 depicts a 4 roll stack transfer coater/laminator used to make the seam tapes of the invention.

Figure 2 depicts a seam tape of this invention. It has a layered construction wherein (20) is the thermoplastic hot melt polyurethane adhesive, (21) is a thin, not necessarily continuous, layer of the cured adhesive prepolymer (22) is the expanded microporous polytetrafluoroethylene with the pores essentially filled with the cured adhesive precursor and (23) is a microporous expanded polytetrafluoroethylene membrane without the precursor in the pores.

Figure 3 depicts another embodiment of this invention wherein (24) is the thermoplastic hot melt polyurethane adhesive layer, (25) is the cured polyurethane prepolymer in (26), which is the microporous expanded polytetrafluoroethylene membrane with the pores substantially filled with the cured polyurethane prepolymer (25).

Figure 4 depicts a third embodiment of this invention wherein (27) represents the thermoplastic hot melt polyurethane adhesive layer, (28) represents the cured polyurethane adhesive, (29) represents the microporous expanded polytetrafluoroethylene layer substantially filled with (28) and (30) is a thin, non-woven fabric.

Figure 5 depicts a two layer tape of the invention applied to the sewn seam of a two layer breathable laminate. The microporous expanded polytetrafluoroethylene layer in the tape is 31. The thermoplastic hot melt polyurethane adhesive (32) has fully penetrated the shell fabric (33) at the seam, down to the microporous expanded polytetrafluoroethylene layer (34) in the fabric laminate. The sewing thread (35) in the seam is completely encapsulated.

It is understood that the thickness of the layers can be varied as desired.

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DETAILED DESCRIPTION OF THE INVENTION

This invention relates to a new construction of tape for sealing sewn seams to prevent liquid water entry in waterproof and breathable garments or drapes. The new sealing tapes are of several structures, all of which utilize an essentially colorless and transparent thermoplastic hot melt adhesive layer joined to a microporous expanded polytetrafluoroethylene layer which is partially filled with a liquid polyurethane prepolymer which is cured to an essentially colorless and transparent elastomeric high melting or non-melting flexible polymer, so leaving a thin layer of exposed microporous expanded polytetrafluoroethylene as one surface of the seam tape. Alternatively, the hot melt layer may be adhered to the microporous expanded polytetrafluoroethylene which is substantially filled with the liquid polyurethane prepolymer and a thin non-woven fabric is adhered to said liquid filled membrane and the liquid prepolymer is solidified and moisture cured to substantially colorless high melting or non-melting, flexible, transparent polymer.

The cured prepolymer should have a melting point well in excess of the melting point of the layer of thermoplastic hot melt polyurethane adhesive to provide an adequately large window for operation of the heat sealing procedure without worry that said cured prepolymer will soften or liquify and allow delamination.

The thermoplastic hot melt adhesive is a polyurethane which melts below 250°C, preferably below 200°C and even more preferably below 180°C, but in all cases above 125°C. The thermoplastic polyurethane layer should have a melt flow rate (as determined by ASTM 1238 under conditions K155/15) of greater than 10g./min. and less than 200g./min.; the preferred range for the melt flow index is greater than 20g./min. and less than 150g./min. to ensure flow into and through the fabric layer of the seam during application.

Further, for two or three layer seam tape, the thermoplastic hot melt polyurethane adhesive layer should be between 1 mil and 6 mils thick. The thickness required in the thermoplastic hot melt adhesive layer is determined by the thickness of any fabric through which the molten adhesive must flow to form a bond and

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seal with that material in the structure being sealed which provides the waterproofing properties. In many waterproof and breathable structures that waterproofing material is microporous expanded polytetrafluoroethylene.

The thin, microporous flexible films or membranes consist of microporous expanded polytetrafluoroethylene which melts significantly higher than the thermoplastic layer. Said membranes are liquid waterproof, but water vapor-permeable, i.e., breathable. The preferred membrane is microporous expanded polytetrafluoroethylene with a thickness of 0.1 mil to about 1.0 mils. More preferably, the expanded microporous polytetrafluoroethylene is 0.2 mils to 0.5 mils thick, has a weight of 2g./m^2 to 5g./m^2 and a porosity of 40% or greater prepared according to the teachings of U.S. Patent 3,953,566 and U.S. Patent 4,187,590.

The curable polyurethane adhesive prepolymer needs, after curing to be transparent and flexible and to form strong adhesive bonds with the thermoplastic hot melt adhesive layer as well as with the non-woven fabric layer, if used. Further, as a prepolymer it must have a sufficiently low viscosity as a liquid to flow into the membrane and when cured it must melt well above the melting point of the thermoplastic layer to prevent delamination. Materials with melt viscosities between 500 cps. and 50,000 cps. or preferably 1000 cp. to 20,000 cps. at 100°C are required. Preferably the melting point of the cured polymer is in excess of 200°C . More preferably, it acts like a thermoset resin and does not melt, but decomposes. Moreover, in the solid form it must be insoluble in water in all cases, and, in some, should be unaffected by dry cleaning solvents.

Liquid polyurethane prepolymers which are useful are essentially linear and may be cured with added curing agents or with atmospherically borne moisture. The most preferred liquid polyurethane prepolymers and polymers are those described in U.S. Patent 4,532,316.

The fabric that is optionally used in the seam tapes herein should be open in structure and be very thin. It can be ~~knitted~~, woven or non-woven polyester or polyamide. It should have a soft

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hand and good drape properties. In the three layer construction, the openness, necessary to ensure that the molten thermoplastic layer can penetrate its structure at seam crossovers can be reflected by its airflow properties. Fabrics should have Gurley numbers below 0.05 sec. and preferably below 0.01 sec. Gurley numbers are the number of seconds required for 100 cc. of air to pass through 6.45 square centimeters of fabric sample. The preferred fabric is a melt bonded, non-woven polyester which is 4.5 mil thick, weighs 3.7g./m^2 and is available from Vertac in Athens, Georgia.

In the case of the two layer tape wherein the pores of the membrane are only partially filled with polymer, there is a distinct layer of expanded polytetrafluoroethylene that has not been penetrated by the polyurethane prepolymer, and this layer of expanded membrane prevents blocking or sticking of the tape to itself. In the case of the three layer tape using the lightweight non-woven fabric for the outer layer, the membrane is essentially filled with polyurethane prepolymer, which acts as the adhesive to bind the non-woven fabric and the thermoplastic polyurethane layer. In the latter case, the non-woven fabric prevents blocking of the tape to itself.

The act of filling the pores of the microporous expanded polytetrafluoroethylene with an transparent essentially colorless polyurethane reduces the number of light reflecting surfaces in the membrane and reduces its opacity. During application, from both the heat and pressure applied in the seam sealing process, the tape becomes even less opaque, being essentially transparent and approaches the optical properties of the polyurethanes.

Even when a layer of thin non-woven fabric is included in the composite, the tapes are remarkably less opaque and less noticeable than the conventional seam sealing tapes made with heavier microporous expanded polytetrafluoroethylene and thermoplastic polyurethanes, but made without filling of the pores of the membrane with prepolymer.

The new tapes are thinner than conventional tapes, and this reduces bulk, provides more suppleness in the finished seams which improves drape. The suppleness and thinness, achieved without

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loss of dimensional stability while being applied hot, also provides for greater ease in forming seals especially where seams meet and one tape must crossover another.

Additionally, the inclusion of the microporous expanded polytetrafluoroethylene membrane provides a barrier to thread breaking through the seam tape, so creating leaks.

The low opacity provides a seam which is very attractive, the tape being almost invisible. And last, the reduced reflectance is a very desirable and demanded property for exterior sealed seams in operating room garments and drapes in which reflection must be reduced as much as possible to reduce glare effects.

Gurley Number Determination

Expanded polytetrafluoroethylene was tested for Gurley Number, defined in this patent as the time in seconds for 100 cc. of air to flow through 6.45 cm^2 of test material under a pressure drop of 1.2 kPa. The test device, a Gurley Densometer Model 4110, was employed in a method similar to Method A of ASTM D726-58. The sample was clamped into the testing device with a reinforcing mesh screen (150 microns) under the test sample to prevent rupture of the test sample.

Bubble Point Determination

Expanded polytetrafluoroethylene was tested for bubble point, defined in this patent as the pressure required to blow the first bubble of air detectable by its rise through a layer of liquid covering the sample. A test device, similar to the one employed in ASTM F316-80, was used consisting of a filter holder, manifold and pressure gauge (maximum gauge pressure of 275.8 kPa). The filter holder consisted of a base, a locking ring, an o-ring seal, support disk and air inlet. The support disk consisted of a 150 micron mesh screen and a perforated metal plate for rigidity. The effective area of the test sample was $8.0 \text{ plus or minus } 0.5 \text{ cm}^2$.

The test sample was mounted on the filter holder and wet with anhydrous methanol until the sample became transparent. The

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support screen was then placed on top of the sample and the top half of the filter holder was tightened in place. Approximately 2 cm. of anhydrous methanol was poured over the test sample. The pressure on the test sample was then gradually and uniformly increased by the operator until the first steady stream of bubbles through the anhydrous methanol were visible. Random bubbles or bubble stream of the outside edges were ignored. The bubble point was read directly from the pressure gauge.

Melt Viscosity

The melt viscosity of the chemical substance was measured between parallel oscillating discs at 100°C. on a Rheometrics System 4 rheometer.

Continuous Wash Test

Samples were washed for 12 hour increments in 18 gal. of cold water without detergents in a Sear Robuck, Kenmore model C110489 washer in accordance with the method of the American Fabric Institute. The washed samples were hung to dry before testing for leakage.

Two layer laminates were first covered with a thin liner fabric before washing to prevent agitator damage to the laminate.

Leak Testing

Washed and dried samples of taped seams were tested for leakage using the Suter test apparatus according to the method described in A.A.T.C.C.-127 using 1 psi pressure for one minute.

EXAMPLES

Materials

A polyurethane prepolymer was prepared from diphenylmethane diisocyanate, polyethylene glycol 1450 and 1, 4-hydroquinone

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di(B-hydroxyethyl) ether by the teaching of U.S. Patent 4,532,316. The prepolymer's viscosity was between 3000 and 10,000 cp. at 100°C.

Porous expanded polytetrafluoroethylene with a thickness of 0.2 mil, a methanol bubble point of 25 psi and a weight of 3.7g./m² was made by the teachings of U.S. Patent 3,953,566.

A 4.5 mil 1/2 oz./sq.yd. thermally bonded non-woven polyester fabric was obtained from Versatec in Athens, GA, as their Grade No. 9316467 which has a Gurley number of 0.0095 sec.

A 6 mil thermoplastic hot melt polyurethane adhesive film was obtained from Bemis Associates in Watertown, MA, and was identified by their part number 6-3205.

The coating and laminating equipment for two and three layer seam tape are illustrated in Figure 1.

In Figure 1, 1 is a gravure roll nipped to a silicone transfer roll (2) which is nipped to a chrome roll (3) which in turn is nipped to a second silicone roll(4). Rolls 1, 2 and 3 are heated. The liquid adhesive prepolymer (9) is applied to the gravure roll via a heated tray and doctor blade (10).

In Figure 1, the porous polytetrafluoroethylene membrane scaffold (5) is coated with the polyurethane prepolymer (9) to form coating (7) which is then married to the thermoplastic hot melt adhesive layer (6) to form a two layer seam tape.

To make a three-layer tape, the second payoff feeds a fabric (11) onto the chrome roll under the microporous expanded polytetrafluoroethylene membrane. The nip pressure causes the liquid prepolymer to flow into and through the membrane pores to adhere the fabric. The membrane filled with liquid prepolymer is then joined with the thermoplastic hot melt adhesive layer and the prepolymer is allowed to solidify and to cure with atmospheric moisture.

Example 1 - Two Layer Seam Tape

For making two layer seam tape, the 4 roll stack shown in Figure 1 is put in line with two payoffs and one take-up. The four roll stack consists of gravure roll (1) engraved with a

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quadrangular pattern, 20 cells/sq.cm. each having a depth of 160 micrometers. The gravure roll is nipped to a 60 Durometer silicone roll (2) which in turn is nipped to a chrome roll (3) which in turn is nipped to a second 60 durometer silicone roll (4). The gravure roll (1), silicone roll (2) and chrome roll (3) are heated at 100°C. The polyurethane prepolymer is fed to the gravure roll via a trough (10) heated at 100°C. In practice, the hot prepolymer was doctored onto the gravure roll (a) which in turn transfers the material (9) to silicone roll (2). The hot liquid prepolymer was then transferred to the porous expanded polytetrafluoroethylene membrane (5) and the coated membrane mated to (7) the thermoplastic hot melt polyurethane adhesive (6) between the chrome roll (3) and the second silicone roll 4. The product 8 was led to a take-up. The product was allowed to cool and solidify and then to cure with atmospherically borne moisture for two days to develop the final seam tape properties.

Example 2 - Three Layer Seam Tape

For a three layer seam tape, the equipment in Figure 1 was used, except that provision was made for feeding non-woven polyester fabric (11) into the nip between the silicone transfer roll (2) and the chrome roll (3), the fabric being against the chrome roll. The polyurethane prepolymer was transferred to the microporous expanded polytetrafluoroethylene membrane and the pressure exerted by the nip between rolls 3 and 4 forces the prepolymer completely through the expanded PTFE membrane to present an adhering surface to the polyester non-woven fabric. Application of the hot melt adhesive was as in Example 1. Again, after take-up, the combined product is allowed to stand for two days to cure the prepolymer with atmospherically borne moisture.

The samples of Examples 1 and 2 were tested for their reflectance properties both before and after application to a polyester fabric (available under the trademark Pongee® fabric) that had been coated with the same breathable polyurethane adhesive prepolymer used in Examples 1 and 2. Sample A is the sample of Example 1. As seen in Table I, its reflectance "over

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white" exhibited a high value. But when Sample A was used to seam seal the coated polyester fabric, the % reflectance values "over white" dropped considerably. This is shown in "Sample B" of Table I and illustrates the highly improved reflectance of a sample of the invention.

For Sample C, Table I shows the reflectance values of a commercially available seam seal tape. This tape is made of microporous polytetrafluoroethylene coated with a polyurethane hot melt adhesive. When it was coated (as in Sample D) on the same coated polyester fabric used for Sample B. The reflectance values did not decrease anywhere nearly as much as they did for Sample B.

Sample E of Table I shows the reflectance values for the tape of Example 2 and illustrates the low reflectance and concomitant transparency of the tape.

Samples B and E were tested for leakage and the tapes did not delaminate or leak.

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TABLE I
REFLECTANCE VALUES (%)¹

Sample		Over White	Over Black	Overall ³ Average
A. Example 1, 6 mil two layer		76.82	19.2	
B. Polyurethane coated R ²		31.8	34.9	
Pongee, sealed with L ²		35.1	34.4	
A (seam) Avg.		33.4	34.7	34.1
C. Commerically available 6 mil, 2 layer "GORE-TEX®" seam tape		83.4	69.2	
D. Polyurethane coated R		60.1	61.6	
Pongee, sealed with L		58.4	59.7	
B Avg		59.3	60.8	60.1
E. Example 2, 6 mil R		40.9	39.7	
layer on polyurethane L		40.3	39.0	
coated pongee Avg.		40.6	39.4	40.0
White Backing		90.2	96.1	
Black Backing		.2	1.5	

1. ASTM D2244

2. R=Right L=Left

3. $\frac{YRW + YRB + YLW + YLB}{4}$

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What We Claim Is:

1. A thin, layered tape for sealing sewn textile seams against liquid entry comprising:

- (a) a first layer of a thermoplastic hot melt adhesive joined to,
- (b) a second layer comprising a thin, microporous membrane whose micropores along at least one surface are filled with a curable, liquid high melting or non-melting, breathable polyurethane adhesive prepolymer, the first layer being in contact with the surface of the second layer containing the filled pores,
- (c) said tape being capable of becoming substantially transparent and colorless with low reflectivity upon application of heat and pressure.

2. The thin layered tape of Claim 1 wherein the membrane is expanded microporous polytetrafluoroethylene.

3. The thin layered tape of Claim 2 wherein the micropores along the other surface are not filled with the curable breathable polyurethane adhesive.

4. A thin layered tape of Claim 2 wherein the micropores of the membrane layer are substantially filled with high melting or non-melting polyurethane and a third layer of thin non-woven fabric is adhesively joined to the second face of said substantially filled membrane with said high melting or non-melting polyurethane.

5. A thin layered tape of Claim 4 wherein the third layer of fabric is comprised of a thin non-woven spun-bonded polyester.

6. The thin layered product of Claim 1, 2, 3, 4 or 5 wherein the thermoplastic hot melt adhesive is a thermoplastic polyurethane which melts between 125°C and 250°C.

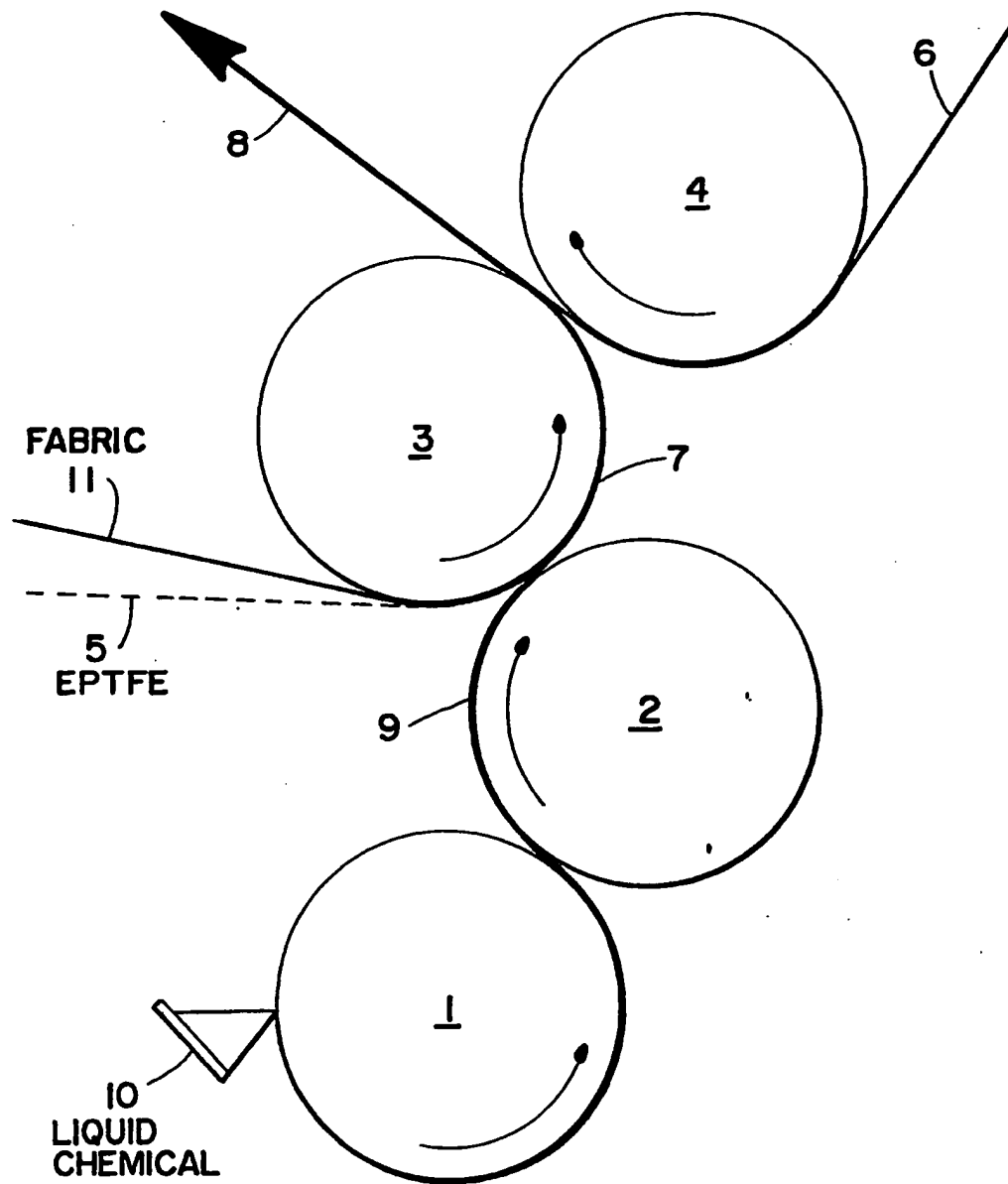
$\frac{1}{2}$ 

Fig. 1

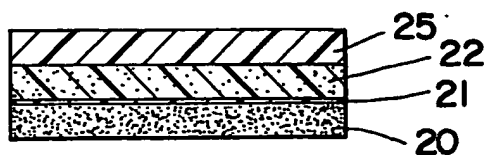
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Fig. 2

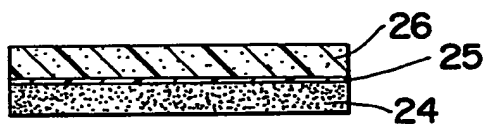


Fig. 3

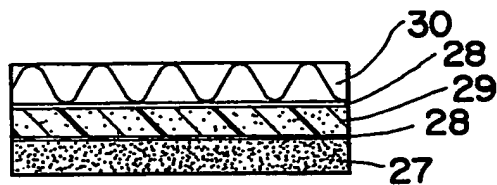


Fig. 4

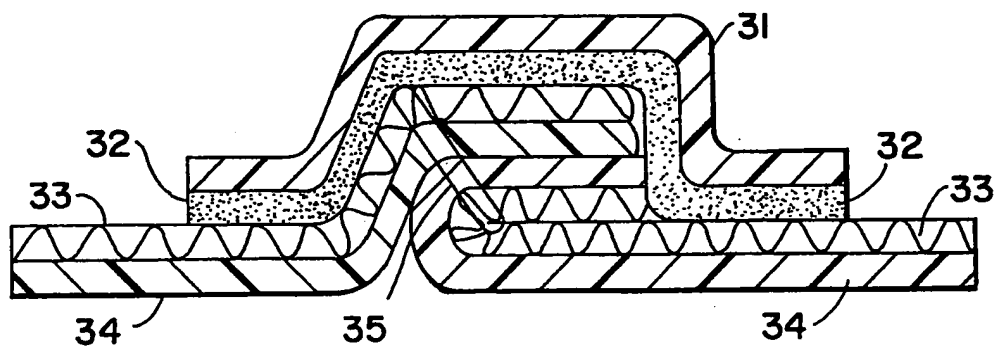



Fig. 5

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 90/06575

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) ⁶		
According to International Patent Classification (IPC) or to both National Classification and IPC		
Int.Cl. 5 B32B5/18 ; B32B27/04 ; C09J7/02 ; //A41D27/24		
II. FIELDS SEARCHED		
Minimum Documentation Searched ⁷		
Classification System	Classification Symbols	
Int.Cl. 5	A41D ; B32B ; C09J	
Documentation Searched other than Minimum Documentation to the extent that such Documents are Included in the Fields Searched ⁸		
III. DOCUMENTS CONSIDERED TO BE RELEVANT⁹		
Category ¹⁰	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
A	US,A,3953566 (GORE) 27 April 1976 see abstract see column 1, lines 1 - 40 see column 16, line 25 - column 17, line 55; claims (cited in the application) ---	1, 2
A	EP,A,0307123 (JAPAN GORE-TEX, INC.) 15 March 1989 see page 5, line 5 - page 8, line 15; claims ---	1
A	US,A,4532316 (HENN) 30 July 1985 see claims 1-21 (cited in the application) ---	3, 4
A	WO,A,8802604 (J.E. BARTASIS ET AL) 21 April 1988 see claims ; figure ---	1
<p>¹⁰ Special categories of cited documents :</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&" document member of the same patent family</p>		
IV. CERTIFICATION		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
21 FEBRUARY 1991	25.03.91	
International Searching Authority EUROPEAN PATENT OFFICE	Signature of Authorized Officer DE JONGE S.J.P. 	

**ANNEX TO THE INTERNATIONAL SEARCH REPORT
ON INTERNATIONAL PATENT APPLICATION NO.**

PCT/US 90/06575
SA 42025

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
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21/02/91

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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		DE-A, B 2123316	02-12-71
		FR-A- 2090775	14-01-72
		GB-A- 1355373	05-06-74
		JP-A, B, C51030277	15-03-76
		JP-A, B 50138387	04-11-75
		NL-A- 7107000	23-11-71
		SE-B- 392582	04-04-77
		US-A- 3962153	08-06-76
		US-A- 4187390	05-02-80
EP-A-0307123	15-03-89	JP-A- 1069341	15-03-89
		AU-B- 600585	16-08-90
		GB-A- 2209707	24-05-89
		US-A- 4916017	10-04-90
US-A-4532316	30-07-85	AU-B- 555571	02-10-86
		AU-A- 4102185	05-12-85
		CA-A- 1236945	17-05-88
		EP-A- 0172610	26-02-86
		GB-A, B 2159526	04-12-85
		JP-A- 61051020	13-03-86
WO-A-8802604	21-04-88	EP-A- 0287643	26-10-88
		JP-T- 1501160	20-04-89
		US-A- 4924525	15-05-90
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